# MR spectroscopy with parabolic magnetic field: spin-oscillator coupling effect

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The spectrum of the spin particle in oscillatory potential subjected to external parabolic magnetic field $\mathbf{B} = (B_0 + Gx + \overline{G}x^2)\hat{\mathbf{z}}$ is obtained. The structure of energy levels of the considered system allows to identify the frequency of the oscillator via the spectrum of spin sublevels coming only from *one* oscillatory level. The effect is due to the gradient terms in the form of the field.

In a recent decade one can observe a rapid grow of interest in the Stern-Gerlach (SG) interaction [1] within MR spectroscopy [2–8]. The main reasons are the following: (i) partial saturation of the possibilities of the conventional MR methods based on Bloch-Purcell paradigm [2] (low sensitivity, especially for the nuclear MR (NMR), growing costs of aparature, existence of cheaper, competitive methods like e.g. ultrasonic ones), (ii) recent reports on new and promising effects (e.g. atomic force microscopy [3,4], MR microimaging [5,6], MR spectroscopy for selected Zeeman states [7,8] (iii) the need for new solutions in the domain of NMR quantum computing (the main problem in the recent implementations of quantum computing is low signal intensity) [9] (iv) the recent development in the domain of ferromagnetics and superconductors allowing to produce strong magnetic fields of required shapes. In this situation one is seeking for new, both theoretical and experimental solutions for MR, in particular by exploiting the SG interaction.

Recently Sidles [4] has proposed the interesting model of spin-$\frac{1}{2}$ particle mounted to the harmonic oscillator. The particle is assumed to interact with the magnetic field via Stern-Gerlach Hamiltonian plus some time-dependent, constant in space magnetic field. He solved the model and showed that the wavepacket of the particle exhibits the splitting into two parts moving in opposite directions. He also predicted the importance of possible oscillator based NMR spectroscopy for molecular imaging devices.

In this Letter we consider the coupled spin-oscillator system in external heterogeneous magnetic field. The model is solved exactly and provides physical spectrum in which the oscillator frequency is coupled to the field derivatives parameters. It implies that in principle one can recognise the frequency of oscillator basing only on signals coming form transitions between sublevels of *one* oscillatory level.

Let us consider the particle that subjects to the simple dynamic due to the harmonic oscillator Hamiltonian $\mathcal{H}^{\Omega}_{osc}(x-a)$. Here $a$ is the position of the minimum of the oscillator potential with respect to the origin of the reference frame (we imagine that the particle is in a sample of finite dimension $l$ situated in the origin of the reference frame, so that $a$ can vary within the region $(-l, l)$). The Hamiltonian of the particle

$$\mathcal{H}^{\Omega}_{osc}(x) = \frac{-\hbar^2}{2m}\frac{d^2}{dx^2} + \frac{m\Omega^2 x^2}{2}. \tag{1}$$

has the following spectrum and corresponding eigenvectors:

$$\mathcal{E}_n = \hbar\Omega(n + \frac{1}{2})$$
$$\Psi_n(\Omega; x) = (\frac{m\Omega}{2^{2n}(n!)^2\hbar\pi})^{\frac{1}{4}} \cdot H_n(\sqrt{\frac{m\Omega}{\hbar}}x) \cdot exp(-\frac{m\Omega}{2\hbar}x^2) \tag{2}$$

with Hermite polynomials $H_n$.

Suppose now that the particle has the spin $S$ and recall that the quantum operator corresponding to the projection of the spin onto the $z$ axis is described as $\hat{\mathbf{I}} = \sum_{M=-S}^{S} \hbar M|M\rangle\langle M|$ with the corresponding spectrum and eigenvectors $\hbar M, |M\rangle$.

Consider the inhomogeneous magnetic field

$$\mathbf{B}(x) \equiv B(x)\hat{\mathbf{z}} = (B_0 + Gx + \bar{G}x^2)\hat{\mathbf{z}} \tag{3}$$

along $\hat{\mathbf{z}}$ axis. Here $G$, $\bar{G}$ stands for the gradient and second derivative parameters. Note that the field has the value $\mathbf{B}_0 = B_0\hat{\mathbf{z}}$ for $x = 0$ an that in general it depends *quadratically* on the spatial coordinate $x$.

Now if we put our particle in the field (3) then its Hamiltonian becomes

$$\mathcal{H} = \mathcal{H}^{\Omega}_{osc}(x-a) \otimes I - \gamma B(x) \otimes \hat{\mathbf{z}}\hat{\mathbf{I}} \tag{4}$$

In the above formula the difference between spatial and spin degrees of freedom has been stressed. In particular the identity operator $I$ has been used. The new energy spectrum of our particle can be found seeking the eigenvectors in the form $|\phi\rangle \otimes |M\rangle$ $(M = -S, -S+1, ..., S)$. This leads effectively to the sequence of $2S+1$ $M$-dependent shifted quantum oscillators. Eigenvalues each of those oscillators can be easily solved and the final energy spectrum of the particle is given by

$$E_{M,n} = \hbar\sqrt{\Omega^2 - \frac{2\gamma\bar{G}\hbar M}{m}}(n+\frac{1}{2}) - \gamma(B_0 + Ga + \bar{G}a^2)\hbar M - \frac{\gamma^2(G+2\bar{G}a)^2\hbar^2M^2}{2m(\Omega^2 - \frac{2\gamma\bar{G}\hbar M}{m})} \tag{5}$$

and the corresponding eigenvectors can be written in the form

$$\Phi_{M,n} = |\phi_{M,n}\rangle \otimes |M\rangle \tag{6}$$

The spatial coordinate function is defined by the oscillator eigenvectors (2) in the following way

$$\phi_{M,n}(x) \equiv \Psi_n(\tilde{\Omega}_M; x - a - \frac{\gamma(G+2\bar{G}a)\hbar^2M}{m\tilde{\Omega}_M^2}) \tag{7}$$

where $\tilde{\Omega}_M \equiv \sqrt{\Omega^2 - \frac{2\gamma\bar{G}\hbar M}{m}}$.

It is important to note that the squared component of the field represented by its second derivative $\bar{G}$ must not be too large as then it suppresses the squared term of the particle oscillator potential $\frac{m\Omega^2q^2}{2}$ leading to Hamiltonian unbounded form below. This occurs for the values violating the inequality

$$\bar{G} < \frac{m\Omega^2}{2\gamma\hbar M_{max}}. \tag{8}$$

where $M_{max} = S$. In such case the first term of the spectrum (5) becomes imaginary for some $M$. Now, if the particle were a light, spin endowed part of molecule (the model which we shall discuss subsequently), this could be interpreted as the dissociation of the molecule caused by the strong gradient of the magnetic field (herewith, we will keep, a bit roughly, the term "dissociation"). The last inequality establishes the boundaries of our model. In this context it is convenient to introduce the new discrete parameter defined by

$$\overline{M} = \frac{2\gamma\overline{G}\hbar M}{\Omega^2 m} \tag{9}$$

Using $\overline{M}$ the inequality (8) writes as

$$\overline{M}_{max} < 1 \tag{10}$$

i.e. the dissociation occurs if $\overline{M}$ reaches the unity. In terms of $\overline{M}$, the dissociation occurs if $\overline{M}$ reaches unity, so that the scope of the presented model is described by inequality $\overline{M} < 1$. Roughly speaking $\overline{M}$ is equal to the ratio of $M$ to the number of the maximal spin level for which dissociation occurs. As we are interested in the effects caused by gradients, we put $B_0 = 0$. Then the energy spectrum can be written in the following form

$$E_{M,n} = \hbar\Omega(\frac{1}{2}+n)\sqrt{1-\overline{M}} - \frac{m\Omega^2(G/\overline{G})a}{2}\frac{\overline{M}}{1-\overline{M}} - \frac{m\Omega^2a^2}{2}\frac{\overline{M}}{1-\overline{M}} - \frac{m\Omega^2(G/\overline{G})^2}{2}\frac{\overline{M}^2}{4(1-\overline{M})} \tag{11}$$

Consider first the case of infinitely small sample concentrated in the origin of the reference frame (i.e. put $a = 0$). Then we obtain especially appealing form of energy spectrum

$$E_{M,n}(a=0) = E^{qu}_{osc}(\Omega, n)\sqrt{1-\overline{M}} - E^{cl}_{osc}(\Omega, G/\overline{G})\frac{\overline{M}^2}{4(1-\overline{M})}, \tag{12}$$

where $E^{qu}_{osc} = \hbar\Omega^2(\frac{1}{2}+n)$ and $E^{cl}_{osc}(\Omega, G/\overline{G}) = \frac{m\Omega^2(G/\overline{G})^2}{2}$.

Here we have two competitive terms: the first one represents the energy of *quantum* oscillator of frequency $\Omega$ (i.e. the energy of the particle in absence of the field) while the second one is nothing but the total energy of the *classical* oscillator of the same frequency, of the amplitude determined by the *shape* of the field. Both terms have weights depending on the scaled spin number. Let us now keep the value of $\overline{G}$ constant (suitably chosen, in order not to be too close to the dissociation regime, e.g. to have $\overline{M} \simeq 0.1$). Now varying the linear gradient $G$ we obtain smooth transition from quantum oscillator regime to the classical one. Provided $G$ is sufficiently large, so that the classical part dominates, we have a kind of *amplification* of the vibrations of the particle by the magnetic field. In both cases the frequency $\Omega$ determines the ratio of splitting of the spin levels. Thus by measuring the latter, we can obtain the information about the value of the frequency. Note that, remarkably, the same is *impossible* in homogeneous field: the possibility of monitoring vibrational frequency via magnetic resonance is exclusively due to the *gradient* terms.

Note that the double "quantum-classical oscillator" structure of the energy levels is due to quadratic gradient term. Indeed, for $\overline{G} = 0$ we we cannot use the scaled spin level number $\overline{M}$ as the latter is defined for $\overline{G} \neq 0$. In such a case we have no "dissociation" as there is no denominator singularity in formula (5). If we put, instead, $G = 0$ (with $a \neq 0$ we still have two oscillators, however the latter one follows from finite dimensions of the sample (so that the centre of the oscillations may feel nonzero value of magnetic field)

$$E_{M,n}(G=0) = E^{qu}_{osc}(\Omega, n)\sqrt{1-\overline{M}} - E^{cl}_{osc}(\Omega, a)\frac{\overline{M}^2}{1-\overline{M}} \tag{13}$$

On Fig. 1 we present these levels for electronic MR (EMR) versus quadratic gradient $\overline{G}$ for the system of spin $S = 3/2$ (we keep $G$ being nonzero, but weak). One can observe remarkable crossing points for sufficiently large $\overline{G}$. Again, for a given value of $\overline{G}$ the level difference is determined by the vibrational frequency $\Omega$.

Let us summarise our results. The very important feature of the obtained spectrum of the particle is that the

spin quantum number $M$ has been *coupled* to the characteristic frequency $\Omega$ of the particle. This means that now for *fixed* quantum oscillator number $n$ the M-dependent energy sublevels form the structure which *depends on the characteristic frequency* $\Omega$ *of the particle*. Indeed, putting $G = \bar{G} = 0$ one can immediately see that it *does not* happen for uniform field $\mathbf{B}(x) = \mathbf{B}_0$. It is remarkable result for the following reasons. Imagine the molecules with spin-less heavy "core" and spin $S$ light part coupled to it. Assume that we have a mixture of *unknown* molecules of that kind, each of them possessing unknown characteristic frequencies $\Omega_1$, $\Omega_2$, ... . Then the fact that the spectrum (5) (with nonzero $G$ or $\bar{G}$) depends on the frequency will provide the possibility to identify them basing directly on the NMR techniques that generically deal with $2S + 1$ spin sublevels of the given level.

It is interesting that if we have $\bar{G} = 0$ and nonzero $G$ then the $n$ dependent term does not depend on quantum number $M$. Of course, the structure of spin sublevels does not depend on $n$. Then all the molecules of one kind (distinguished by $\Omega$) would give *the same* signal from $M$ sublevel of *all* oscillatory levels. A very interesting feature of the considered interaction is the emerging amplification of the vibrational mode to the form of classical oscillator. The effect is due to nonzero $\overline{G}$. The amplitude is determined by the shape of the field, and the amplification is obtained for high values of the linear gradient.

Concerning the experimental realiation of the presented spin-oscillator coupling effect note that for the typical molecular frequences ($\approx 10^{14} Hz$) one would need enormously strong gradients to observe the effect. Indeed, the strongest gradients obtained so far amount to $10^6 T/m$ [4]. Now, for molecule with incorporated atom of oxygene the above gradients allow to realize the effect for frequences up to $10^4 Hz$. Besides the highest frequency for which the effect can be observed satisfies roughly $\overline{G} \sim \Omega^2$. On the other hand, there exist molecular systems that exhibit low frequences [10]. Thus the effect is difficult but still possible to realize experimentally.

In conclusion, we have demonstrated that MR spectroscopy with parabolic magnetic field provides subtle spin-oscillator coupling effects, that allow to select the characteristic frequency of the system. It seems to open new prospects for the MR applications such as quantum level separations [7,8] or quantum computation [9]. Finally, we expect that the idea of incorporating higher order gradients will prove fruitful in looking for new solutions within MR domain. We also believe that the obtained results will stimulate development of techniques of producing strong gradient field of different shapes.

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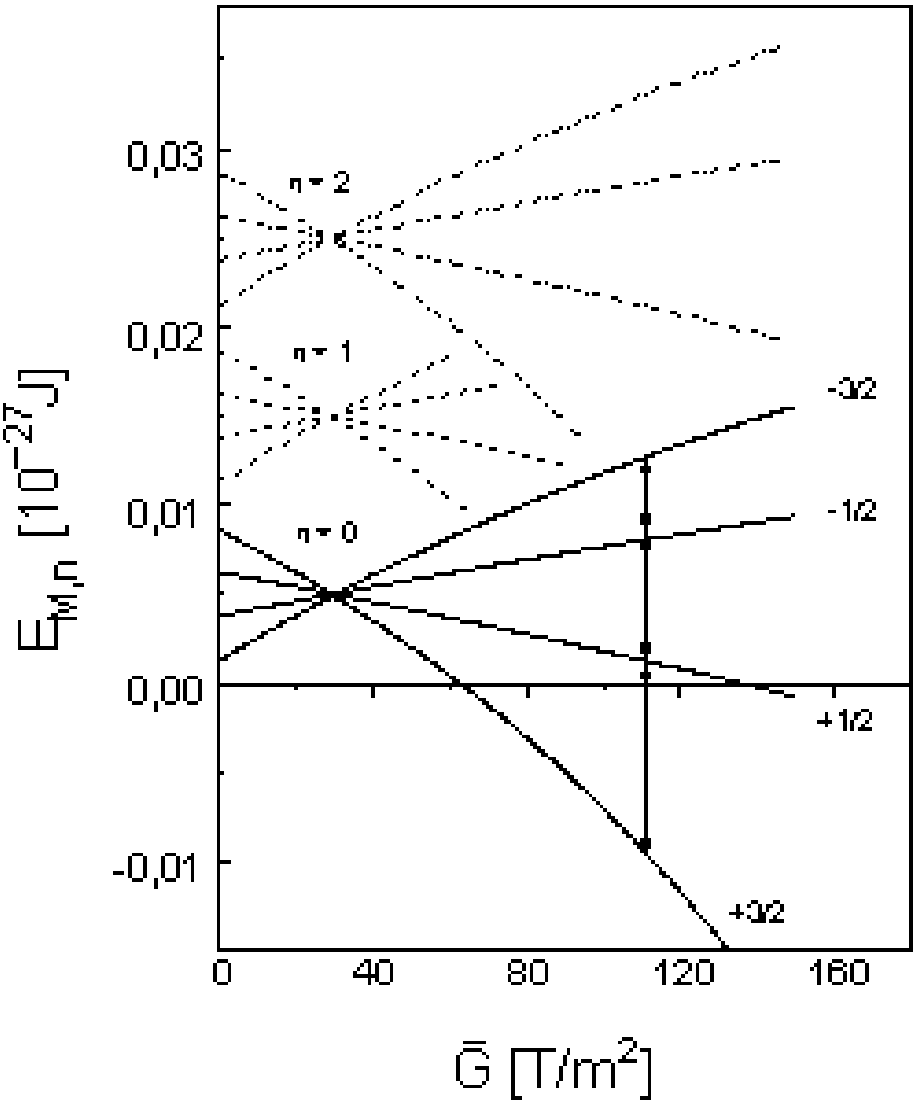
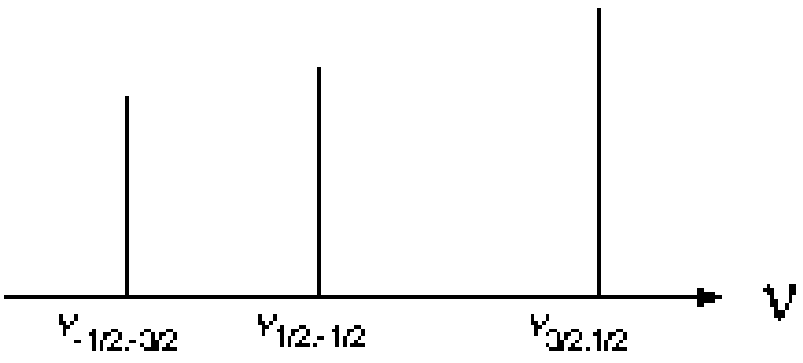
FIG. 1. Structure of the EMR energy levels (a) in parabolic field for finite dimension of the sample ($S = 3/2$, $a = 10^{-4} m$, $\Omega = 10^5 Hz$, $G = -0.003 T/m$) and oscillatory spectrum (b).

a)


0,03
0,02
0,01
0,00
-0,01
n = 2
n = 1
n = 0
-3/2
-1/2
+1/2
+3/2
0
40
80
120
160
$E_{M,n}$ [$10^{-27}$ J]
$\bar{G}$ [T/m$^2$]

b)


$\nu_{-1/2,-3/2}$
$\nu_{1/2,-1/2}$
$\nu_{3/2,1/2}$
$\nu$